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Project No. G-41-X87
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FINAL REPORT

Submitted to the
Division of Materials Sciences
Department of Energy
Washington DC 20545

SUBMITTED BY: Andrew Zangwill
Georgia Institute of Technology
Atlanta, GA 30332

TITLE: Kinetics of Heteroepitaxy

CONTRACT #: DE-FG02-97ER45658

GRANT PERIOD: 9/1/00-8/31/02

TOTAL AMOUNT: $130,000

OVERVIEW

Most of the funds for this two-year grant were used to support Dr. Max Petersen, a post-doctoral fellow who joined my group in February 2000. He is a now a senior researcher at Accelrys, Inc., a scientific simulation concern. The remainder of the funds provided summer support for the PI. This lead to a collaboration with a Georgia Tech colleague, Prof. Michael Schatz, and two physicists at the Weizmann Institute. No Georgia Tech graduate students were supported on this grant. Our activities were reported in:

1. ***Ripples Settle Surface Behavior***, A. Zangwill, Physics World, September 2000, pp. 23-24.

2. ***Convective Instability of Strained Step-Flow Growth***, N. Israeli, D. Kandel, M. Schatz, and A. Zangwill, Surface Science **494,** L735 (2001).

3. ***Level Set Approach to Reversible Epitxial Growth***, M. Petersen, C. Ratsch, R.E. Caflisch, and A. Zangwill, Physical Review E **64**, 061602 (2001).

4. ***Advances in Aggregation***, A. Zangwill, Nature **411**, 651 (2001)

5. *Homoepitaxial* ***Ostwald Ripening***, M. Petersen, A. Zangwill, and C. Ratsch, Surface Science **536**, 55 (2003).

DETAILS

I begin with Papers 3 and 5 above, which represent the core of the research. Our interest was to extend the so-called ``level-set'' method [1] to simulate **reversible** epitaxial growth. This is the situation where atoms are forbidden to detach from step edges.

To model irreversible growth, the level set method solves the diffusion equation on every terrace (with absorbing boundary conditions) to find the monomer density $c(\mathbf{r})$. For each island step edge, the growth velocity is

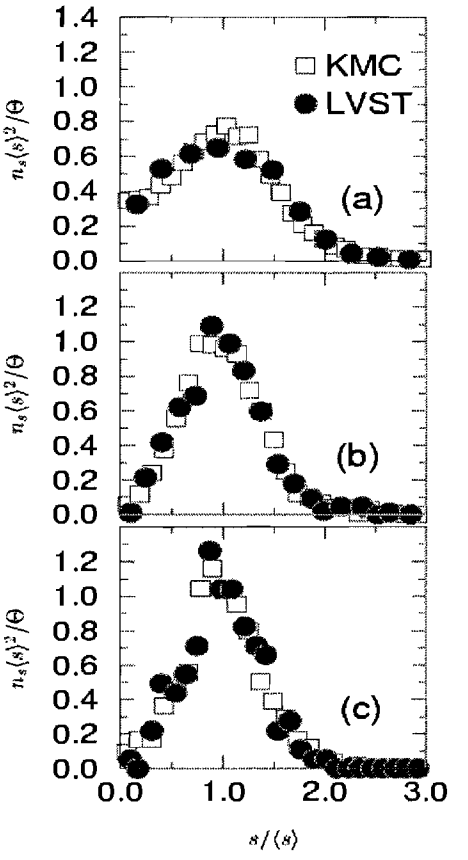
$$v_+ = -a^2 D \left. \frac{\partial c}{\partial n} \right|_{\text{step}}$$

where a is the lattice constant and D is the terrace diffusion constant. The key to the method is the algorithm used to track the motion of each terrace. Island nucleation is treated as a stochastic process

Dr. Petersen's task was to generalize the level set (LVST) method to handle reversible growth—the situation where any atom can detach at will from any step edge—in a manner that does not create additional computational overhead. This is an absolute prerequisite to any practical level set approach to heteroepitaxy. He was completely successful, albeit after a very long period of code development and debugging. There are (at least) two ways to take account of detachment. One alters the boundary condition on the diffusion equation [2]. This considerably complicates the level set formalism. As an alternative, we supplemented the growth velocity above with a "detachment velocity"

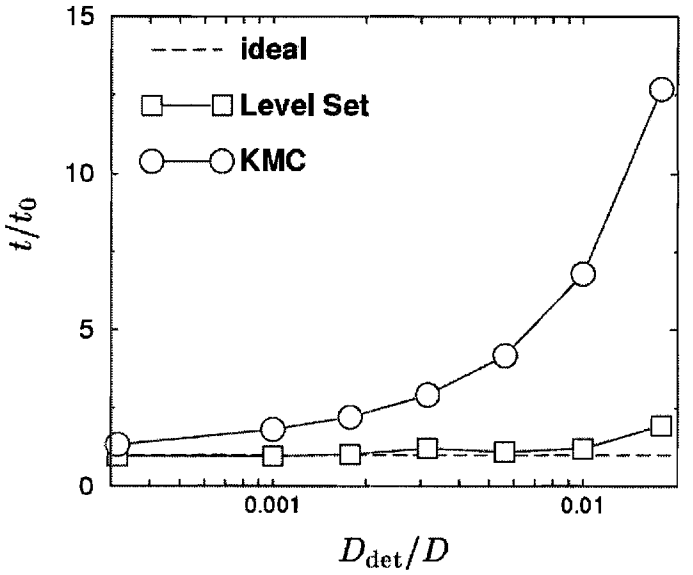
$$v_- = a^2 D_{\text{det}} p_{\text{esc}} \lambda.$$

Here, D_{det} is the detachment rate from an edge, p_{esc} is the probability that a detaching atom actually escapes from the island, and λ is the linear density of singly coordinated edge atoms. It is also necessary to decide where to put the detached atoms and to invent a stochastic algorithm to deal with very small islands that disintegrate due to detachment. When all this is done, we find that the submonolayer island size distribution computed by LVST agrees quantitatively with the corresponding KMC distribution (see figure at right). From top to bottom, the three panels have $D_{\text{det}}/D = 10^{-4}, 5\times10^{-3}$, and 10^{-3}.



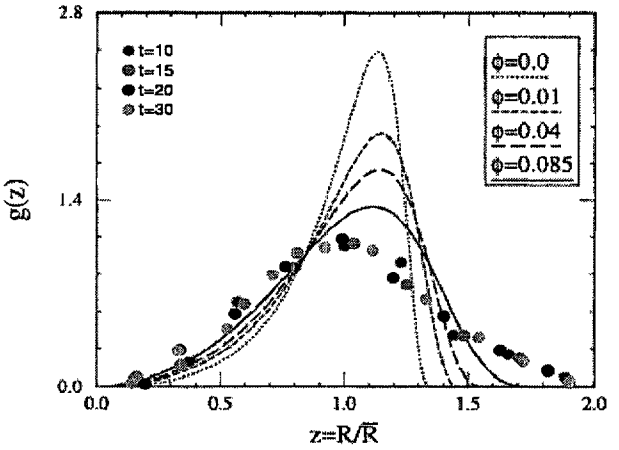
This was encouraging, but not decisive until we compare the computation time needed for the LVST simulation with the time required to

perform the KMC simulations. This is shown in the figure below.



As suggested above, the CPU time for a KMC simulation increases rapidly as the detachment rate increases. The LVST simulation suffers negligibly in comparison. From our earlier KMC simulations of 3D island formation, we know that a value of $D_{\mathrm{det}}/D = 0.01$ is typical for realistic systems and temperatures. The graph above tells us that the corresponding LVST simulation (not optimized) would run at least five times faster.

To further check the correctness of our methodology, we elected to study the problem of Ostwald ripening for 2D epitaxial islands. That is, how does the "as-grown" island size distribution (depicted earlier) change when the system is held at temperature after the deposition flux is turned off? This is a subject of considerable experimental interest [3]. It was also studied not long ago using KMC simulations [4]. Oddly, the latter authors did not compare their simulated island size distributions with the current best analytic theory of ripening in two dimensions [5]. This mean field theory predicts that the distribution approaches an asymptotic (late time) form that depends only on the coverage. The figure below shows that our higher coverage results ($\phi = 0.25$) follow the trend of the analytic theory very well. Note that the data from different annealing times (dots) all collapse onto one curve.



Finally, a casual lunch conversation with a Georgia Tech colleague—an experimental fluid dynamics expert—led us to re-analyze a model of strain-induced step-bunching that was introduced a few years ago [6]. The original study began with a uniform array of steps in step-flow motion and asked the usual question: does a periodic perturbation of step separations grow or decay as growth proceeds? We asked a question that is more typical in the field of hydrodynamics: does a localized perturbation spread more rapidly than is propagates (absolute instability) or does it propagate more rapidly than is spreads (convective instability)? The distinction is important because a convectively unstable system is sensitive to subsequent perturbations while an absolutely unstable system is not. This has implications for morphological patterning at the nanoscale.

As reported in Paper 2 above, we discovered that the model we studied is indeed convectively unstable toward step bunching over a very wide range of material parameters and growth conditions. The most interesting consequence of this fact arises if we supplement the uniform growth flux with a highly collimated "pencil" of atoms that can be scanned across the surface to perturb the step pattern locally. On can also imagine using the "shadow" cast by a scanning tunneling microscope to accomplish the same thing (with reverse contrast). When this raster is used to "write" a single pattern on the flowing steps, the convective nature of the growth causes "echoes" of the pattern to appear periodically on the surface. Non-linear effects eventually become important, but our calculations suggest that the echoes reproduce the original pattern may times with surprising good

fidelity. To our knowledge, this is an entirely novel way to generate lateral pattering in an otherwise featureless step-flow situation.

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